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Recommendations to Improve Wildlife Exposure Estimation for Development of Soil Screening and Cleanup Values

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EDITOR'S NOTE

This paper represents 1 of 6 articles generated from a workshop entitled "Ecological soil levels: next steps in the development of metal clean-up values" (September 2012, Sundance, Utah, USA). The purpose of the workshop was to provide managers and decision makers of contaminated sites in North America with appropriate methods for developing soil clean-up values that are protective of ecological resources. The workshop focused on metals and other inorganic contaminants because of their ubiquity at contaminated sites and because their natural occurrence makes it difficult to determine adverse levels.

ABSTRACT

An integral component in the development of media-specific values for the ecological risk assessment of chemicals is the derivation of safe levels of exposure for wildlife. Although the derivation and subsequent application of these values can be used for screening purposes, there is a need to identify the threshold for effects when making remedial decisions during site-specific assessments. Methods for evaluation of wildlife exposure are included in the US Environmental Protection Agency (USEPA) ecological soil screening levels (Eco-SSLs), registration, evaluation, authorization, and restriction of chemicals (REACH), and other risk-based soil assessment approaches. The goal of these approaches is to ensure that soil-associated contaminants do not pose a risk to wildlife that directly ingest soil, or to species that may be exposed to contaminants that persist in the food chain. These approaches incorporate broad assumptions in the exposure and effects assessments and in the risk characterization process. Consequently, thresholds for concluding risk are frequently very low with conclusions of risk possible when soil metal concentrations fall in the range of natural background. A workshop held in September, 2012 evaluated existing methods and explored recent science about factors to consider when establishing appropriate remedial goals for concentrations of metals in soils. A Foodweb Exposure Workgroup was organized to evaluate methods for quantifying exposure of wildlife to soil-associated metals through soil and food consumption and to provide recommendations for the development of ecological soil cleanup values (Eco-SCVs) that are both practical and scientifically defensible. The specific goals of this article are to review the current practices for quantifying exposure of wildlife to soil-associated contaminants via bioaccumulation and trophic transfer, to identify potential opportunities for refining and improving these exposure estimates, and finally, to make recommendations for application of these improved models to the development of site-specific remedial goals protective of wildlife. Although the focus is on metals contamination, many of the methods and tools discussed are also applicable to organic contaminants. The conclusion of this workgroup was that existing exposure estimation models are generally appropriate when fully expanded and that methods are generally available to develop more robust site-specific exposure estimates. Improved realism in site-specific wildlife Eco-SCVs could be achieved by obtaining more realistic estimates for diet composition, bioaccumulation, bioavailability and/or bioaccessibility, soil ingestion, spatial aspects of exposure, and target organ exposure. These components of wildlife exposure estimation should be developed on a site-, species-, and analyte-specific basis to the extent that the expense for their derivation is justified by the value they add to Eco-SCV development. Integr Environ Assess Manag 2014;10:372–387. © 2013 The Authors. Integrated Environmental Assessment and Management Published by SETAC

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INTRODUCTION

An integral component in the development of media-specific values for the ecological risk assessment of chemicals is the derivation of safe levels of exposure for wildlife, defined here as birds and mammals. Although the derivation and subsequent application of these values can be used for screening purposes, there is a need to identify the threshold for effects when making remedial decisions during site-specific assessments. Methods for

evaluation of wildlife exposure are included in the US Environmental Protection Agency (USEPA) ecological soil screening levels (Eco-SSLs) (USEPA 2005a), registration, evaluation, authorization, and restriction of chemicals (REACH) (EC 2006) and other risk-based soil assessment approaches (Fishwick 2004; CCME 2006). The goal of these approaches is to ensure that soil-associated contaminants do not pose a risk to avian and mammalian species that directly ingest soil, or to species that may be exposed to contaminants that persist in the food chain (i.e., in plants, invertebrates, or prey of higher predators). Both the Eco-SSL approach and the REACH approach are intended to protect all (or, in practice, 95%) species, and they are not designed to address site-specific concerns for evaluating effects to the most at-risk wildlife at specific sites. The Eco-SSL approach is intended to be a component of screening-level ecological risk assessments (SLERAs), whereas the REACH approach is intended as a generic assessment of the risk presented by the ongoing production and use of specific substances in a broader chemicals management framework. Both approaches incorporate broad assumptions in their overall scope, the exposure and effects assessments, and in the risk characterization process. The outcome of using these broad assumptions is often that the threshold for concluding risk is very low, and conclusions of risk may be made for soil metal concentrations that fall in the range of natural background. In terms of identifying areas where remediation is necessary, this approach is far from practical because it makes it difficult to exclude sites from detailed consideration as a result of the screening assessment.

A workshop was held in September, 2012 that evaluated existing methods and explored recent science about factors to consider when establishing appropriate remedial goals for concentrations of metals in soils. A Foodweb Exposure Workgroup was organized to evaluate available methods for quantifying exposure of wildlife to soil-associated metals through soil and food consumption and to provide recommendations for the development of ecological soil cleanup values (Eco-SCVs) that are both practical and scientifically defensible. The specific goals of this article are to review the current practices for quantifying exposure of wildlife to soil-associated contaminants via bioaccumulation and trophic transfer, to identify potential opportunities for refining and improving these exposure estimates, and finally, to make recommendations for application of these improved models to the development of site-specific remedial goals protective of wildlife. Although the focus is on metals contamination, many of the methods and tools discussed are also applicable to organic contaminants.

EXPOSURE ESTIMATION IN THE USEPA ECO-SSLS AND UNDER REACH

Methods and assumptions used by the USEPA Eco-SSLs and under REACH were evaluated as a starting point for identifying limitations of current exposure estimation approaches. These limitations were then used as the basis for discussion of potential improvements to exposure estimation methods.

USEPA wildlife Eco-SSLs

The USEPA Eco-SSLs for wildlife are soil concentrations associated with an exposure dose that is equal to a no observed adverse effect level (NOAEL) (USEPA 2005a). Eco-SSLs were developed for use in the screening-level ecological risk assessment phase of the 8-step process for assessing ecological risks at Superfund sites (USEPA 1997). The Superfund process allows for the refinement of screening levels (e.g., Eco-SSLs) through the use of more realistic (i.e., less conservative) assumptions before the baseline ecological risk assessment (BERA) problem formulation step. Eco-SSLs have been developed for 16 metals and/or metalloids and 5 organic contaminants for both birds and mammals. Multiple conservative assumptions are intentionally used to take account of the uncertainties associated with the assessment. The results, therefore, represent the concentrations of contaminants in soil that are believed to be protective of individual birds or mammals that come into contact with soil or ingests biota that live in soil (USEPA 2005a).

In the practice of risk assessment for a given site, maximum contaminant concentrations from soil samples collected at a Superfund site are compared to Eco-SSL concentrations. If the maximum concentration does not exceed the Eco-SSL, then it can be confidently concluded that the contaminant in question does not present an unacceptable risk to the considered bird or mammal receptor, which are selected to provide examples typical of the major potential exposure pathways. However, if the maximum concentration exceeds the Eco-SSL, the contaminant in question may present a risk and should be retained for additional evaluation in the baseline ecological risk assessment. It should be noted that due to their conservative nature, simply exceeding an Eco-SSL should not be taken as an indicator of risk. Rather, it simply indicates that the available data are insufficient to conclude that risks are absent and that additional evaluation is necessary. Furthermore, Eco-SSL guidance states that Eco-SSLs are not intended to be used as remedial goals.

In their simplest form, wildlife Eco-SSLs are derived by solving:

$$HQ = \frac{\text{Exposure Dose}}{\text{TRV}};$$

where HQ (hazard quotient) = 1, TRV (toxicity reference value) represents a NOAEL dose extracted from published literature, the exposure dose is the amount of a contaminant in the diet that is taken up by the organism from consumed food or directly from ingested soil. Both the exposure dose and TRV are expressed in the same units (mg contaminant/kg body weight/day).

The fully expanded model for Eco-SSL derivation is:

$$HQ_j = \frac{\left(\left[Soil_j \times P_s \times FIR \times AF_{sj}\right] + \left[\sum_{i=1}^{N} B_{ij} \times P_i \times AF_{ij}\right]\right) \times AUF}{TRV_j}$$

where HQ_j = hazard quotient for contaminant (j) (unitless); $Soil_j$ = concentration of contaminant; (j) (mg/kg dry weight); N = number of different biota types in the diet; B_{ij} = concentration of contaminant (j) in diet type (i) (mg/kg dry weight); P_i = proportion of diet type (i) in diet; P_s = soil ingestion as proportion of diet; FIR = food ingestion rate (kg food [dry weight]/kg body weight [wet weight]); AF_{ij} = absorbed fraction of contaminant (j) from biota type (i); AF_{sj} = absorbed fraction of contaminant (j) from soil (s); TRV_j = toxicity reference value for contaminant (j) (mg/kg/day); and AUF = area use factor.

It is not feasible to calculate Eco-SSLs for all bird and mammal receptors that may possibly be exposed at a contaminated site. Eco-SSLs were therefore calculated for

bird and mammal species selected as representatives of major trophic groups (although exposure estimation was specific to each trophic group, each relied on the same TRV). These are herbivores, insectivores, and carnivores. Surrogate bird and mammal species for which Eco-SSLs were derived were selected to conservatively represent their respective trophic groups, sharing general characteristics associated with potential vulnerability, such as small body size to maximize metabolism and food ingestion and a small home range to maximize site AUF. Other criteria used to guide surrogate species selection included the presence of a clear direct or indirect exposure pathway link to soil, foraging focused in terrestrial, upland habitats, and diet composition that could be simplistically classified (USEPA 2005a). Species selected as surrogates for Eco-SSL derivation are all common and widely distributed in the United States and included meadow vole (Microtus pennsylvanicus), short-tailed shrew (Blarina brevicauda), long-tailed weasel (Mustela frenata), mourning dove (Zenaida macroura), American woodcock (Scolopax minor), and red-tailed hawk (Buteo jamaicensis).

Because Eco-SSLs are applied in the screening-level phase, several simplifying assumptions are made in their calculation. Absorbed fractions of contaminants from diet and soil (AF_{ij} and AF_{sj}) are both assumed to be 100% (e.g., contaminants in food and soil are 100% bioavailable for uptake by the consumer). Similarly, it is assumed that individual birds and mammals represented by the Eco-SSLs reside and forage exclusively on a contaminated site (i.e., AUF¼1). All diets were assumed to consist exclusively of a single food type; 100% plant foliage for herbivores, 100% earthworms for insectivores, and 100% small mammals for carnivores.

In addition to these assumptions, the Eco-SSL wildlife exposure model was parameterized using exposure factors based on the selected surrogate species. Food ingestion rates (FIR) for each surrogate are represented by the arithmetic mean of high-end estimates (i.e., 90th percentile) for food ingestion reported in USEPA (1993) or other published species-specific studies. The proportion of soil in the diet (P_s) for each species is represented by the 90th percentile of the soil ingestion rate calculated probabilistically using the model from Beyer et al. (1994). Concentrations of contaminants in diet items (B_{ij}) were calculated using analyte–biota specific regression models (if available) or median bioaccumulation factors (in the absence of a regression model). The regression model is favored as it captures changes in bioconcentration factor that can occur when organisms are exposed at different soil concentrations (see below).

Limitations to the USEPA Eco-SSLs. Many of the simplifying assumptions that ensure that the wildlife Eco-SSLs are conservative and suitable as screening values also limit their realism, and therefore their applicability within higher tier assessment frameworks. These limitations need to be considered in the development of Eco-SCVs. For example Eco-SSLs only address selected species whose diets are simplified to a single food type. This underrepresents the complexity of wildlife communities that may be present at contaminated sites, the breadth of food resources that individual wildlife species may consume and be exposed to, and the variability in the extent and mobility of metal bioaccumulation among different food resources. Because most wildlife species consume a variety of food types, with diet composition frequently varying by age class, sex, and season, accurate estimation of dietary exposure requires a more detailed integration of diet composition than is included in the Eco-SSLs.

Similarly, although a growing body of literature indicates that the bioavailable fraction of contaminants in ingested soil, sediment, plant tissue, and animal tissue is less than 100% and likely much lower than that for contaminant forms used in TRV studies, the Eco-SSLs assume 100% bioavailability. Incorporating measured bioavailability data or estimates of bioavailability (e.g., bioaccessibility) could decrease the estimated exposure dose, and thereby influence the outcome of assessments. For example, using a waterfowl physiologically based extraction test, Turner and Hambling (2012) estimated that the bioaccessibility of metals to the mute swan associated with sediment and a range of food types was a fraction of the total metal concentrations associated with these matrices. In fact, the highest observed bioaccessibility was approximately 13% (for Zn from a mixture of food types). The bioaccessibilities for the majority of food types measured for Ni and Pb were less than 1%. Given that bioaccessibility is a conservative (high) estimate of the true absorbed dose (i.e., bioavailability), these results underscore the point that the bioaccessibility and bioavailability of metals through diet borne exposure can be very low, and that this information can greatly decrease the estimated exposure dose (e.g., by a factor that could vary from between 10-fold and up to 100-fold).

Another limitation to the Eco-SSLs is the manner in which they are calculated. Eco-SSLs are calculated deterministically, with values for all parameters represented by either mid-range (i.e., bioaccumulation factors [BAFs]) or high-end (i.e., FIR and P_s) point estimates from their respective distributions (USEPA 2005a). Although this is conservative and suitable for screening-level evaluations, it does not take into account the underlying variability of each parameter in the exposure model, nor does it account for correlations among parameters. Exposure models, such as those used in the Eco-SSLs can readily be implemented probabilistically such that distributions of exposure are generated that may then be used to generate a distribution of estimated exposure for comparison to the TRVs, either as a dose–response distribution or a distribution of TRVs. This comparison produces estimates of the likelihood that contamination present at the site will result in exposures that could produce adverse effects, a more useful metric for risk evaluation and remedial decision making. Sensitivity analyses (Cullen and Frey 1999) of the model may also be carried out to identify those parameters that exert the greatest influence on the final exposure distribution. For example, probabilistic implementation of exposure models using site-specific data was used as part of the BERA for the Coeur d'Alene River (CdA) Basin (USEPA 2001) for species identified as highly exposed (spotted sandpiper [Actitis macularius], tundra swan [Cygnus columbianus], and vagrant shrew [Sorex vagrans]) based on deterministic exposure estimation. Using this probabilistic analysis, the proportion of assessment areas within the CdA Basin with soil or sediment concentrations sufficiently high to produce adverse effects for each species was identified. In addition, sensitivity analyses indicated which model parameters were most influential (i.e., Pb concentrations in soil and/or sediment for sandpipers, soil ingestion for swans, and arthropod bioaccumulation for shrews).

A related limitation is that Eco-SSLs do not address spatial aspects of exposure. Both birds and mammals are mobile, moving about their environment in pursuit of food, water, and shelter to a greater or lesser extent depending on the species.

They experience exposure in various portions of the home range based on the time spent and the behavior they engage in (i.e., foraging, drinking, loafing, sleeping, etc.) in each area. The Eco-SSL exposure model includes a simplistic parameter (AUF) to account for the fraction of time an animal spends on a contaminated site. However, in a screening level risk assessment, it is assumed that receptor animals reside exclusively at a contaminated site, and so the AUF is set to 1. This approach neither accounts for receptor movements, nor the underlying spatial variability of contamination on site. Use of more complex models, such as the Spatially Explicit Exposure Model (SEEM) (Wickwire et al. 2004) or Eco_SpaCE (Loos et al. 2010) are needed to more fully address the spatial aspects of dietary exposure.

A further common criticism expressed in relation to Eco-SSLs derivation for wildlife, and indeed other soil quality criteria derived from laboratory toxicity data, has been that values can be set at close to (or even below) background concentrations. A recent development designed to ground-truth the validity of laboratory data derived environmental quality criteria has been to use multiple site data sets that include information on community composition and pollutant concentrations to derive "field" species sensitivity distributions. These values are derived using abundance data to identify relationships between species occurrence and pollutant concentrations from which critical concentrations relating to the loss of a given percentage of species can be derived. To date, studies of this type have largely been conducted to validate existing limits (for stream and sediment communities) rather deriving alternatives (Leung et al. 2005; Stockdale et al. 2010; Struijs et al. 2011). To apply a similar approach for wildlife, challenges related to greater range size, high species mobility, and variation in metal bioavailability due to heterogeneous metals species composition within an animals foraging range, provide complications not yet dealt with in the existing studies. Further metapopulations of wildlife at polluted sites may be maintained by immigration, or alternatively, offsite foraging may reduce the potential to exceed a toxic dose. For these reasons, analysis of community structure for quality criteria derivation or ground truthing for wildlife could prove a challenging task that will be hard to satisfactorily address. Consideration of these limitations serves to illustrate that the basic equations used for screening-level wildlife exposure assessments, such as wildlife Eco-SSL model, represent a foundation on which more detailed wildlife exposure models can be built. These simple equations can be refined to describe site-specific conditions and circumstances in cases where the SLERA is insufficient for drawing risk management conclusions. For example, as part of the BERA for the CdA River Basin (USEPA 2001), dietary exposure models were developed and evaluated for 42 wildlife receptors (24 birds and 18 mammals). In addition to the large number of species and broad diversity of feeding guilds represented, the exposure models for the CdA BERA included measures of contaminant bioaccessibility in soil and diet, based on both site-specific and literature-derived data. The models were also implemented probabilistically (for some highly exposed species) to gain a more holistic view of uncertainty associated with potential exposure.

REACH

The REACH regulation requires that all chemical substances that are produced in or imported into Europe at volumes greater than 1000 tons/yr be registered with the European Chemicals Agency (ECHA). A main component of the registration process is the Chemical Safety Report (CSR), which obligates the registrant to demonstrate safety for each designated use of the registered substance for a range of human health endpoints and exposure scenarios (e.g., occupational and consumer health), as well as environmental processes and ecotoxicological endpoints.

Bioaccumulation is one of the environmental processes for which data are required in the CSR. The approach by which the consequences of bioaccumulation is assessed within REACH is called "secondary poisoning." Secondary poisoning essentially addresses concerns associated with "toxic effects in the higher members of the food chain…which result from ingestion of organisms from lower trophic levels that contain accumulated substances."

The REACH approach is generic, and the REACH guidance addresses only one terrestrial food chain, which is soil–earthworm–worm-eating birds or mammals. The estimation of the exposure dose is therefore focused on the estimation of bioaccumulation by earthworms and incidental soil ingestion by worm-eating birds and mammals.

Bioaccumulation factors are used to quantify earthworm tissue concentrations using the following relationship:

$$C_{\text{earthworm; dry wt}} = C_{\text{soil; dry wt}} \times BAF_{\text{dry wt}}.$$

Unlike the Eco-SSL approach, the REACH approach does not recognize the dependence of BAFs on soil concentration (McGeer et al. 2003). Use of a constant BAF, therefore, represents a simplifying assumption in the assessment when compared to the United States approach. An additional limitation that is shared with the Eco-SSL approach is that the influence of soil chemistry on bioaccumulation potential is not included as part of the assessment. There is ample evidence that soil properties, other than metal concentrations, can influence trace metal accumulation in plants and animals. For example, DeForest et al. (2011) observed an inverse relationship between Ni BAFs for earthworms and soil Ni concentrations, as well as between BAFs and soil cation exchange capacity (CEC). Similarly, Pauget et al. (2012) found that inclusion of pH and CEC into regression models allowed a significantly better prediction of Cd and Pb concentrations in snail tissue than those derived using metal concentration alone. These results highlight the importance of considering soil factors when predicting tissue concentrations, although they are currently not included. In REACH, the median (50th percentile) BAF alone is used to estimate earthworm tissue concentrations.

The exposure term within the secondary poisoning risk characterization is referred to as the Predicted Environmental Concentration–oral, or PEC_{oral}. The PEC_{oral} is simply defined as $C_{earthworm}$. Worst-case assumptions are used when other factors that contribute to actual internal exposure dose (e.g., bioavailability and dietary composition are considered). For example, metals associated with earthworm tissue and with soil in the earthworm gut are assumed to be absorbed with 100% efficiency. Likewise, the worm-eating birds and mammals are assumed to ingest only earthworms. As outlined above in relation to Eco-SSL derivation, these are likely to be unrealistic assumptions that need to be carefully considered in the context of refining the current screening-level risk assessment for site-specific use.

In practice, the outcome of the secondary poisoning assessment is used to determine if the operational conditions

of individual registrants of chemical substances meet the definition of safe use. Safe use is defined simply as conditions where the PEC_{oral} is below the Predicted No Effect Concentration–oral ($PNEC_{oral}$). Given the number and influence of default assumptions used in the calculation of each of these terms, safe use is infrequently demonstrated for metal substances. This has led to the refinement of selected risk assessments through modification of the default assumptions, an example of which is presented in the next section.

The source of many of the limitations associated with applying the secondary poisoning assessment to metals is that the guidance was developed with organic contaminants in mind. Metal-specific guidance has been developed (ECHA 2008), and this guidance recognizes many of the steps that DeForest et al. (2011) used in their refinement of the Ni secondary poisoning assessment.

RELATIVE IMPORTANCE OF PARAMETERS IN EXPOSURE DOSE MODELS

Both the Eco-SSL and the REACH exposure dose models use default assumptions for many parameters. Sensitivity analyses were conducted to evaluate how the application of conservative default values influences model output and to quantify the relative influence of individual parameters in these exposure models, including TRV selection, food or soil ingestion, bioaccumulation models, and bioavailability and/or bioaccessibility. The outcome of these analyses can be useful in displaying the relative value of collecting additional information to reduce uncertainties in evaluating exposure and risks from metals to wildlife. This information may be used to improve the derivation of guideline values within the context of both regulatory frameworks and also for assessing the relative values of collecting additional information for use in site-specific risk assessment conducted to support site management decisions.

Sensitivity analyses were carried out on the models for the avian Eco-SSL for Pb and the REACH secondary poisoning assessment for Ni. The avian Pb Eco-SSL was selected because, unlike the plant, soil invertebrate, and mammalian Pb Eco-SSLs, the avian Eco-SSL value for Pb is lower than the 50th percentile for reported background concentrations in eastern and western US soils (USEPA 2005b). The Ni REACH analysis for the terrestrial mammalian food chain was similarly selected because the use of default parameters results in conclusions of risk for soil concentrations that are below reported ambient concentrations for Ni in European soils.

The sensitivity analysis for Pb was conducted by varying the value of the TRV, food ingestion rate (FIR), and the proportion of soil in the diet (P_s) based on ranges reported in USEPA (2005a) and USEPA (2007a) (Table 1). Bioaccumulation was varied by calculating the upper and lower 95% prediction intervals for the earthworm regression models from Sample et al. (1999). Absorbed fractions of Pb from soil (AF_s) and diet (AF_i) were based on in vitro bioaccessibility data from Kaufman et al. (2007). Results from the sensitivity analysis were compared to the distribution of Pb concentrations in background soils from USEPA (2007b).

Selection of the TRV had the greatest influence on the resulting Eco-SSL (Figure 1). The selected NOAEL TRV is the key factor resulting in the low Eco-SSL for Pb and avian species. Bioaccumulation and food ingestion rate were the next most important parameters. The fraction of Pb absorbed from soil and diet, and surprisingly soil ingestion rate, which was varied from 1% to 20%, exerted the least influence. These estimates also fell within the range of background Pb concentrations in North America.

The sensitivity analysis for Ni followed the REACH guidance for terrestrial secondary poisoning and used the mammalian food chain. A soil Ni concentration of 30 mg Ni/kg dry soil was used in the determination of the PEC_{oral} concentration, which is comprised of earthworm tissue plus ingested soil. This concentration (30 mg Ni/kg) represents ambient background concentrations for several soil types (agricultural, grassland, forest) in the European Union (EU) (ECB 2008). The sensitivity analysis was carried out by varying the Assessment Factor (an uncertainty factor applied as part of the REACH process) used in the calculation of the $PNEC_{oral}$, the selection of an alternative $PNEC_{oral}$ value, the bioavailability of Ni associated with the ingested earthworm and soil in the earthworm gut, and the dietary composition. The ranges of these parameters were based on an evaluation of terrestrial mammalian food webs within the EU (DeForest et al. 2011), and are explained in detail in the Supplemental Data. The objective of the analysis was to see which parameters were the most important in reducing the PEC:PNEC ratio below 1, which should be the case for a soil containing background Ni concentrations.

Modifying the Assessment Factor that is placed on the lowest available NOEC concentration of birds and mammals from 30 to 10 had little influence on the PEC:PNEC ratio (Table 2). An alternative $PNEC_{oral}$ of 5.1 mg/kg was calculated, based on the geometric mean of mammalian growth and reproduction toxicity data from the Eco-SSL of Ni. Using this approach, the PEC:PNEC ratio was below 1. However, the aggregation of toxicity data in this way may not be supported in the REACH regulation, given the differences in species, endpoints, and test protocols that are reflected in the database. In this situation, modifications to the exposure dose may need to be considered if this can be supported by studies published in the scientific literature. By using a weighted relative absorption factor (RAF) of 0.036 to account for the bioavailability of Ni associated with earthworm tissues and soil within the earthworm gut for worm-eating mammals, the PEC:PNEC ratio was reduced by a factor of 27. Information for worm-eating birds suggest that the absorption of Ni from food sources by some birds should also be extremely low ($<1\%$) (Turner and Hambling 2012). Accounting for relevant dietary composition had less effect, and the greatest reduction in PEC:PNEC ratio was achieved by using a combination of relevant dietary composition (i.e., 31% earthworms and 69% isopods) and the accompanying bioavailability for each food type that was used (Table 2).

Results of these 2 sensitivity analyses vary, with the Pb analysis showing the importance of the effect concentration used (TRV), and the Ni analysis showing the importance of exposure dose estimation. These results indicate that the evaluation of factors that can modify both effects and exposure need to be evaluated on a metal by metal basis and also according to the relevant food chain.

MODELING CONSIDERATIONS TO IMPROVE EXPOSURE ESTIMATION

The depth of scientific knowledge for the parameters identified in the sensitivity analyses are evaluated in the following sections, with the ultimate objective of identifying refinements that can be implemented now, and those for which additional information is required.

Table 1. Sumary of parameter values selected for sensitivity analysis for the avian lead EcoSSL

Parameter	Value	Units	Basis	Reference
TRV	0.194	mg/kg/d	Lowest NOAEL for reproduction, growth, or survival	USEPA 2005b
	1.63		Eco-SSL NOAEL; highest bounded NOAEL lower than lowest bounded LOAEL for reproduction, growth, or survival	
	196		Highest NOAEL for reproduction, growth, or survival	
FIR	0.017	g/g/d	Lowest value for woodcock	USEPA 2007a
	0.214		Woodcock FIR used for Eco-SSL	
	0.229		Highest value for woodcock	
Ps	0	Proportion of FIR	Lowest value for woodcock	USEPA 2007a
	0.164		Woodcock Ps used for Eco-SSL	
	0.201		Highest value for woodcock	
AFs	0.11	Percent	Minimum percent Pb bioaccessible in soil from firing range based on extraction simulating American Robin digestive physiology	Kaufman et al. 2007
	0.53		Mean percent Pb bioaccessible in soil from firing range based on extraction simulating American Robin digestive physiology	
	0.91		Maximum percent Pb bioaccessible in soil from firing range based on extraction simulating American Robin digestive physiology	
AFi	0.49	Percent	Minimum percent Pb bioaccessible in earthworms from firing range based on extraction simulating American Robin digestive physiology	Kaufman et al. 2007
	0.73		Mean percent Pb bioaccessible in earthworms from firing range based on extraction simulating American Robin digestive physiology	
	0.87		Maximum percent Pb bioaccessible in earthworms from firing range based on extraction simulating American Robin digestive physiology	

Diet composition

Both the Eco-SSL and REACH approaches for assessing the potential for food chain transfer rely on analysis using relatively simple assumptions concerning food chain structure. In the Eco-SSL approach, diets are assumed to consist exclusively of a single food type. Under REACH, simple food chains are again the focus for the secondary poisoning assessment. Although simplifying food chains helps to reduce complexity in the exposure models, it does not represent ecological reality, where diet composition may vary among individuals, gender, age classes, habitats, season, and geography.

Voles, for example, have diets that are comprised of a variety of grass and broadleaf plant species, as well as fungal fruiting bodies and even some insects (Faber and Ma 1986; Abt and Bock 1998; Wheeler 2005). The choice of which plant tissue to consume may affect the extent of exposure, with fruiting bodies and seeds frequently containing relatively low concentrations when compared to leaves and roots (Sinha et al. 2006). Dietary analysis indicating that herbivores like voles ingest fungal bodies and invertebrates may provide a greater source of error for exposure estimation than any variation driven by plant species and/or plant tissue selection. Some fungal species are known to hyperaccumulate metals into fruiting bodies and this is often observed among common species (Rudawska and Leski 2005; Melgar et al. 2009). Consumption of such metal rich fungal structures may, therefore, represent a considerable enrichment of intake above the amounts derived from vegetation only diet. Similarly, inclusion of invertebrates in the diet may also increase exposure levels, because these taxa all assimilate a range of trace metals (Hopkin 1989). The contributions of these dietary items suggests that field assessment for herbivores may reflect an increase in the potential transfer of metals due to the cosmopolitan nature of real diets.

Earthworms are assumed to be the sole invertebrate prey of insectivores in both the Eco-SSL and REACH approaches. Because they are common in many ecosystems and also relatively large organisms, with adult Lumbricus terrestris weighing up to 10 g (Lakhani and Satchell 1970; Kammenga et al. 2003), earthworms are certainly an important food resource. Nonetheless, the assumption of 100% of earthworm tissue in the diet is almost always an over simplification. Diet

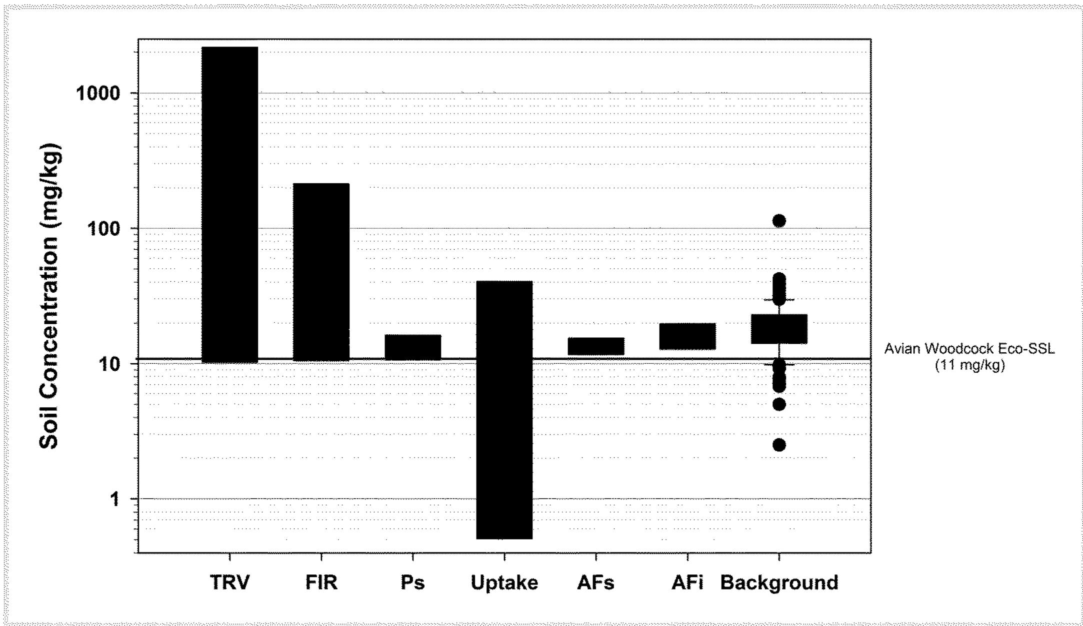

Figure 1. Summary of sensitivity analyses for avian Eco-SSL for Pb compared to the distribution of background Pb concentrations in North America from USEPA (2007b).

generally depends on the species as well as seasonal, climatic, biological, and regional factors. Some species such as the European badger (*Meles meles*) (Cleary et al. 2011), red fox (*Vulpes vulpes*) (Jefferie 1974), and shrew species such as *Sorex*, *Neomys*, and *Blarina* (Churchfield and Rychlik 2006; Whitaker and Ruckdeschel 2006), as well as a number of bird species including thrushes (Gruar et al. 2003) and some predatory birds (Hounsome et al. 2004; Schipper et al. 2012) are only facultative earthworm predators, consuming earthworms only at certain times, such as when they are present on the soil surface (e.g., during wet periods). Other species, such as the European mole (*Talpa europaea*) and birds like the American woodcock (the Eco-SSL receptor) feed predominantly on earthworms, with earthworms constituting greater than 75% of the diet (Hoodless and Hirons 2007).

Prey selection can also represent an important exposure estimation variable for carnivores. For example, it is commonly accepted that predatory birds may favor herbivore and generalist prey species, such as voles and wood mice, above insectivorous species such as shrews. When smaller prey are swallowed intact, the whole body metal concentration is the most accurate estimate of exposure (although some tissues, particularly bone, may not be assimilated). Alternatively, when larger prey are consumed, the predator may preferentially feed on specific tissues, including liver and kidney. This preferential feeding may deliver a high exposure because liver and kidney are repositories of some metals, such as such as Cd and Hg, in mammals (Shore and Douben 1994; Veltman et al. 2007). Avoidance of these tissues in favor of other organs or muscle can have the effect of reducing actual exposure. Accounting for the potential variability in exposure that is associated with prey selection in a practical way could be accomplished in a tiered approach, with Eco-SSLs used as the first tier. A range of typical predator–prey scenarios could be developed for the second tier, allowing the risk assessor to choose the most relevant scenarios for the site in question. If necessary, a third tier would involve

Table 2. Sensitivity analysis of the terrestrial worm-eating mammal food chain. Methodology and other parameters taken from DeForest et al. (2011)

Scenario	AF	Dietborne bioavailability (%)	Dietary Composition	PEC_{oral} to $PNEC_{oral}$ ratio
Default	30	100	100% Earthworm	6.08
Assessment Factor	10	100	100% Earthworm	2.02
Alternative $PNEC_{oral}$	30	100	100% Earthworm	0.87
Bioavailability	30	3.61	100% Earthworm	0.22
Dietary Composition	30	100	30% Earthworm, 70% Arthropod[b]	2.49
Bioavailability and dietary composition	30	3.6[a], 2.5[c]	30% Earthworm, 70% Arthropod	0.08

[a]Relative absorption factor of earthworm tissue plus ingested soil. DeForest et al. 2011
[b]Relevant dietary composition for European shrew. DeForest et al. 2011
[c]Relative absorption factor for arthropods. DeForest et al. 2011.

sampling at the site to validate the outcome of chosen predator–prey scenarios.

As a result of the known variability in diet composition, it is recommended that exposure estimation as a component of eco-SCV development represent site-specific conditions to the extent possible. Field data should be collected to quantify the composition and variability of diets of target receptors. Modeling should address dietary variation such that the most limiting exposure (diet that results in greatest exposure) is identified as this may drive remediation. Time-weighting of exposure may also be considered, if a diet that produces high exposure occurs for only a short duration or during a specific season. Exposures that are maximized during specific periods of the year lend themselves to a seasonal assessment, where the maximum exposure is compared to a TRV that is appropriate for the community structure and life stages of wildlife that are present on the site during the period of maximum exposure. These measurements of exposure are consistent with the recommendations for the choice of TRVs that are used (Mayfield and Fairbrother 2013). For example, short term, idiosyncratic exposures should be compared with acute TRVs. Additionally, if feeding behaviors include preferential consumption of specific tissues, this should be integrated into exposure estimation.

Bioaccumulation models

Metal concentrations in soil are recognized as an important driver of metal accumulation in plants, invertebrates and small mammals. In general, inorganic forms of metals associated with soils are not considered to biomagnify (USEPA 2007d), and therefore the scope of using bioaccumulation models is limited to predictions of metal concentrations in prey items. Extensive field data have shown that biota exposed to higher metal concentrations generally accumulate higher tissue metal concentrations. However, bioaccumulation is not usually directly proportional. Rather as the absolute tissue concentration increases the bioaccumulation factor (i.e., tissue concentration–soil concentration), generally decreases (McGeer et al. 2003). This pattern occurs because bioaccumulation is a nonlinear process with uptake higher at low concentrations, decreasing at higher concentrations due to saturation or toxicity limiting accumulation (Sample et al. 1999). This continuous, nonlinear nature of bioaccumulation is already captured in the Eco-SSLs through the use of empirical field-based soil–biota bioaccumulation models. This contrasts with the REACH secondary poisoning assessment that is based on generic BCF values. Data on metal concentrations in tissues of many major dietary items (i.e., plant foliage, earthworms, small mammals) have been available in the published literature, supporting the development of reliable regression models. Even when data are scarce, multi-element detection methods are rapidly filling this data gap. Although tissue concentration data are becoming increasingly available, synoptically collected soil data, which are required to calculate bioaccumulation models, generally are not. Therefore, it is highly recommended that any site-specific risk assessment collect soil colocated with the tissues that are of interest.

The wide availability of such data on tissue metal accumulation and the relative ease with which site-specific information on tissue accumulation can be obtained through on-site sample collection followed by laboratory analysis can provide a sound basis from which to derive exposure estimates from food items. Ideally, for detailed site assessments, colocated soil and biota samples that represent the prey and/or food of the bird and mammal receptors of interest should be collected. To be consistent with existing published data, total (strong acid extraction) analyses should be conducted on both soil and tissue, although other extractions may also be carried out to evaluate bioaccessibility (see below). The data that are generated from such site-specific monitoring provide the basis for the derivation of site-specific relationships that describe the relationship between soil and biota metal concentrations. Because bioaccumulation is a continuous, nonlinear process, the log linear regression models that are derived can be used as the preferred means to quantify site-specific bioaccumulation relationships. BAFs and/or BCFs should be used only when regression analyses cannot be reliably be determined. Because BAFs and/or BCFs for metals are inversely related to metal exposure concentrations (McGeer et al. 2003), the BAFs and/or BCFs used in the exposure estimation should be consistent with the range of exposures at the site in question. In cases where practical issues (costs, site access, etc.) prevent direct data collection, an alternative approach is to use the literature-derived field-based bioaccumulation models of soil to biota accumulation to estimate trophic transfer.

Bioavailability and/or bioaccessibility

USEPA (2007c) defines bioavailability as "the fraction of an ingested dose that crosses the gastrointestinal epithelium and becomes available for distribution to internal target tissues organs." A related term is bioaccessibility, which is defined as the fraction of the chemical extractable from its matrix (e.g., soil, sediment, food) into the gastrointestinal tract that is available for absorption (Ruby et al. 1996; National Research Council 2003; Koch and Reimer 2012). Although the full Eco-SSL wildlife exposure model includes parameters that address contaminant bioavailability and/or bioaccessibility (e.g., absorbed fractions of contaminants from diet and soil [AF_{ij} and AF_{sj}]), these parameters are assumed to be 100% in the calculation of Eco-SSLs or in the approach for REACH secondary poisoning assessment. Improvement of the estimation of dietary exposure for birds and mammals requires the development of bioaccessibility or bioavailability data and its application in exposure modeling.

Such data may be developed in vivo or in vitro. In vivo evaluation involves administration of metal contaminated soil or food via gavage to the wildlife receptor in question or to an appropriate surrogate species. The absorbed fraction of metals that is determined in such an exposure is referred to as the absolute bioavailable fraction (USEPA 2007c). To place absolute bioavailability into a risk assessment context, the absolute bioavailability of the metal form used to derive the TRV also needs to be derived. Because it is not possible or practical to directly measure the toxicity of metals associated with soils and foods that are found at a specific site, the ratio of the absolute bioavailabilities of the matrix in question (e.g., soil or food) to the substance used in TRV calculations (e.g., a metal salt) can be determined to calculate relative bioavailability (RBA). Provided that RBA was calculated using the same metal salt that serves as the basis for the TRV, it can be used to modify the AF term in the Eco-SSL equation.

Some evidence is available to suggest that absolute bioavailability from in vivo measurements is less than estimates obtained from in vitro bioaccessibility extractions. Vasiluk et al. (2011) compared several in vitro bioaccessibility measurements of soil-associated Ni with absolute bioavailability from in vivo

administrations to rats using the same soils. In general, the absolute bioavailability of Ni from soil was substantially lower than estimates obtained from in vitro bioaccessibility extractions. For example, gastric physiologically based extraction tests (PBETs) for a specific size fraction (<<70mm) from one soil ("PC") extracted 13.5% of the soil-associated Ni, whereas the absolute bioavailability from this soil was less than 1%. Other size fractions, e.g., 150 to 250mm, showed very similar results between the in vivo and in vitro techniques. Absolute bioavailability of $NiSO_4$ was also determined, which allowed the calculation of an RBA that can be applied to risk assessments that use TRVs determined from this substance.

Because in vivo measurements are expensive and conditional in nature, in vitro bioaccessibility techniques are used much more frequently. Koch and Reimer (2012) provide an extensive review of current of in vitro bioaccessibility extraction methods, frequently referred to as PBETs that have been developed for the study of contaminants in soil, sediment, and biological matrices. Many of these methods are based on extraction protocols used in pharmaceutical and nutrition fields. In general, these in vitro methods consist of mixing aliquots of the selected ingested matrix (soil, sediment, or food) with a solution of salts and enzymes whose composition and pH are intended to simulate the conditions within the GI tract of a specific species. This mixture is then maintained at a specified temperature for a specified time (again based on digestion processes for a specific species), after which a sample of solute is extracted for analysis. Total analyses (generally based on concentrated acid digestions) of soil or food are carried out concurrently. Concentrations obtained from the PBET analyses are compared to the total analyses results to quantify the bioaccessible percentage of the contaminant of interest in the selected matrix.

Although one of the first bioaccessibility methods developed (Ruby et al. 1993) was based on rabbit physiology, most subsequent work, both in terms of methods development and in terms of evaluation of analyte- or location-specific bioaccessibility, have focused on human health (Koch and Reimer 2012). These models may be used to support ecological risk assessments (Ollson et al. 2009); however, they are most suitable for a subset of mammalian receptors (those with simple monogastric digestive physiology) and may be sources of considerable uncertainty due to the diversity of digestive physiology among mammalian taxa.

Most bioaccessibility research on metals has focused largely on Pb and As (Koch and Reimer 2012). Some data on other analytes, however, are available, including Sb (Denys et al. 2008), Se (Funes-Collado et al. 2011), Cd and Zn (Roussel et al. 2010), Hg (Zagury et al. 2009), and Pt group metals (e.g., Pt, Pd, and Rh) (Colombo et al. 2008).

The focus of most PBET development has been on contaminant bioaccessibility from soils or sediment. Only recently have studies evaluated bioaccessibility of contaminants in food. Intawongse and Dean (2008) evaluated bioaccessibility of Cr, Cd, Cu, Fe, Mn, Mo, Ni, Pb, and Zn in lettuce and spinach foliage, and in carrot and radish roots. Plants were grown in soil spiked at 3 concentration levels, and then aged for 2 weeks before planting. After reaching maturity, the plants were harvested and analyzed for metal content. The plant samples were extracted using an in vitro gastrointestinal approach to simulate the human digestive tract. Bioaccessibility varied by element, plant species, and type of tissue.

More recently, PBET models have been adapted for evaluating bioaccessibility to ecological receptors. Kaufman et al. (2007) evaluated bioaccessibility of Pb in soil and dietary components at a firing range in Canada. Extractions were performed to simulate gastric digestion of 2 mammal (eastern cottontail [Sylvilagus floridanus] and short-tailed shrew [Blarina brevicauda]) and one bird (American robin [Turdus migratorius]) species that were relevant to the exposure scenario under consideration. Analyses of total (based on nitric and/or hydrochloric acid digestion) and bioaccessible (based on simulated gastric digestion) Pb were performed on sieved soil (<2 mm), unwashed plant foliage, and depurated earthworms. Mean Pb bioaccessibility from earthworm tissue was greater than that for soil, regardless of whether the mammal or bird extraction was considered (Table 3). This may reflect the different speciation states of Pb in earthworm tissue as compared to the mineral present in contaminated soils. In contrast, mean Pb bioaccessibility from vegetation was somewhat lower than that in soil, but variability was much greater.

Moriarty et al. (2012) evaluated the bioavailability of As in soil and insect prey of the masked shrew (Sorex cinereus) at an abandoned gold mine in Nova Scotia. Bioaccessibility was evaluated using a method modified from Ruby et al. (1996) to better represent shrew digestive physiology. Total As in insects was determined following nitric acid digestion and in soils after aqua regia digestion. Similar to Pb, mean As bioavailability from insect prey was greater than that from soil (Table 3).

In an evaluation of bioavailability of As in soil and vegetation to deer mice (Peromyscus maniculatus) at abandoned gold mines in the Northwest Territories, Ollsen et al. (2009) observed that bioaccessibility varied based on the nature and magnitude of contamination. Bioaccessibility was evaluated using a method modified from Ruby et al. (1996) that was designed to represent bioaccessibility to humans. The human model was assumed to be representative for deer mice. Total As in both soil and plant samples was determined after acid digestion. Whereas mean bioaccessibility in vegetation exceeded that measured in soils from the mine site and nearby contaminated forests, mean bioaccessibility from background soils exceeded that of vegetation collected from background (Table 3). The reason for this difference is not discussed by the authors but may be associated with differing forms of As between mine-contaminated sites and background, or the variation in concentration ranges among sites. Arsenic concentrations in the soils varied significantly among the sites with mean concentrations at the mine and nearby forest being 1740 mg/kg and 392 mg/kg, respectively, and 104 mg/kg in background soils (Ollson et al. 2009).

Furman et al. (2006) developed a PBET procedure to evaluate Pb bioavailability to waterfowl exposed to mine-impacted soils in the CdA River basin. They applied their method to field-collected soils that had been sieved (<1 mm) and amended with P, either in the field or in the laboratory, to reduce Pb bioavailability, and also to unamended soils. Whereas mean Pb bioaccessibility in unamended soil was 17.5%, mean Pb bioaccessibility was reduced to 0.52% after P amendment (Table 3). The Pb bioaccessibility results were found to be significantly correlated with absolute bioavailability as measured in mallard ducks (Anas platyrhynchos) fed Pb-contaminated CdA soils.

Turner and Hambling (2012) applied the waterfowl PBET procedure developed by Furman et al. (2006) to evaluate the bioaccessibility of metals in sediment, food, and antifouling paints to mute swans (Cygnus olor). Bioaccessibility from

Table 3. Summary of selected bioaccessibility values for assorted metals in soil/sediment and tissue

Analyte	Taxa represented by PBET extraction: Class	Species	Media extracted	Origin of media	Observed bioaccessibility (%): Mean	Minimum	Maximum	Reference
Lead	mammal	eastern cottontail/ short-tailed shrew	soil	Firing Range	66%	17%	100%	Kaufmann et al. 2007
			earthworm		77%	52%	100%	
			vegetation		50%	8%	160%	
	bird	American robin	soil		53%	11%	91%	
			earthworm		73%	49%	87%	
Arsenic	mammal	masked shrew	soil	abandoned gold mine	11%	2%	19%	Moriarty et al. 2012
			insects		47%	44%	48%	
Arsenic	mammal	deer mice[a]	soil	Mine-tailings	10%	1%	33%	Ollson et al. 2009
			vegetation		30%	15%	44%	
			soil	Mine-forest	20%	17%	22%	
			vegetation		42%	25%	56%	
			soil	Background	23%	7%	29%	
			vegetation		18%	14%	23%	
Lead	bird	waterfowl	soil	mine-impacted soil - no P amendment	18%	12%	28%	Furman et al. 2006
				mine-impacted soil - P amended	0.52%	0.29%	1.04%	
Chromium	bird	waterfowl	sed	Estuary with multiple mining/industry sources[b]	0.24%	0.23%	0.24%	Turner and Hambling 2012
			plant		1.34%	1.21%	1.47%	
Copper	bird	waterfowl	sed		1.42%	0.95%	3.52%	
			plant		2.23%	0.95%	3.52%	
Nickel	bird	waterfowl	sed		0.43%	0.29%	0.57%	
			plant		0.16%	0.14%	0.17%	
Lead	bird	waterfowl	sed		0.46%	0.23%	0.69%	
			plant		0.14%	0.01%	0.26%	
Zinc	bird	waterfowl	sed		1.13%	0.23%	2.03%	
			plant		1.40%	0.10%	2.70%	

[a]Although focus of study was deer mice, the bioaccessibility method used was based on human digestive physiology.
[b]Combined gizzard and intestine bioaccessibility.

sediment was low for all metals; less than 1% for Cr, Ni, and Pb, and less than 5% for Cu and Zn. Bioaccessibility of Cu, Ni, Pb, and Zn from food was similar to or less than that in sediment. Although Cr was the only metal evaluated that displayed greater bioaccessibility in algae as compared to sediment, bioaccessibility was still low (<2%). PBET analyses of sediment and algae with antifouling paint particles resulted in higher bioaccessibility measurements, especially for Cu and Zn. However, mean bioaccessibility following inclusion of up to 10% antifouling paint particles was less than 10% for Cu and less than 15% for Zn.

The literature described above clearly shows that metals bioaccessibility vary by metal, the chemical form of metals (that is in turn influenced by the source, by soil parameters, and by aging processes), and the extraction model used. The development of information on the processes that influence bioaccessibility and bioavailability to wildlife is at its early stages. It is therefore not possible to provide medium-specific default values to use in the calculation of exposure dose. This is consistent with guidance for using bioaccessibility and/or bioavailability for human health risk assessment (USEPA 2007c), which is at a more advanced state of development compared with wildlife applications. Nonetheless, inclusion of site-specific measurement of bioaccessibility is recommended as a component of eco-SCV development to better estimate the diet borne bioavailability of metals associated with soils at the

site of interest, as this is the best approach to address the wide range of bioavailabilities that occur among sites. This recommendation is consistent with that of McLaughlin et al. (2000) for performing weak salt extractions to estimate metal availability to soil organisms (e.g., plants, microbes, and invertebrates) in assessments of contaminated sites. The extraction approach is recommended because metal availability to soil organisms will also vary considerably among sites as a function of the chemical form of the metal and its solubility, and because other available approaches (e.g., empirical leaching and/or aging models proposed by Smolders et al. [2009]) are based on soils spiked with soluble metal salts as opposed to soils that are contaminated by different industrial processes or that have undergone aging in the field. The choice of bioaccessibility methods for a given site should be based on the bird and mammal taxa of interest at the site and should include bioaccessibility analyses of not just soil, but also of the key food types on which these receptors rely.

Soil ingestion

Most birds and mammals ingest soil either inadvertently while foraging (i.e., insectivores ingesting soil adhering to or contained within worms; herbivores consuming soil adhering to roots or deposited on foliage), through grooming, or purposefully to meet nutrient requirements (Suter et al. 2000). Soil ingestion may therefore be a significant exposure pathway at metals contaminated sites. Unlike food or water ingestion, for which allometric models are available with which to estimate ingestion rates (Calder and Braun 1983; Nagy 2001), estimation of soil ingestion in an ecological risk assessment context is limited to empirical measurements.

Although some other researchers have reported soil ingestion rates for some species (i.e., black-tailed jackrabbit [Lepus californicus] and pronghorn antelope [Antilocapra Americana] [Arthur and Gates 1988]; mule deer [Odocoileus hemionus] [Arthur and Alldredge 1979]; cotton rat [Sigmodon hispidus] [Garten 1980]; and domestic dog [Canis familiaris] [Calabrese and Stanek 1995]), the most widely referenced source of soil ingestion rates is Beyer et al. (1994). This work estimated the fraction of soil (P_s) in the diets of 2 reptile, 15 mammal, and 11 bird species by measuring the ash content of diet and scat and calculating P_s using the following model:

$$P_s = (b - y + a \cdot y)/(a \cdot y - c + b);$$

where P_s = proportion of soil in diet (g soil/g dry mass); a = digestibility of food (g absorbed/g dry mass ingested); b = concentration of acid-insoluble ash in food (g/g dry mass); c = concentration of acid-insoluble ash in soil (g/g dry mass); and y = concentration of acid-insoluble ash in scat (g/g dry mass).

Subsequent to Beyer et al. (1994), additional studies on soil and/or sediment ingestion in additional waterfowl species have been published (Beyer et al. 1997, 1999, 2008; Beyer, Audet, et al. 1998; Beyer, Day et al. 1998).

Soil ingestion data are generally lacking for many species, as is information on how soil ingestion varies within species over time or in relation to habitat and diet composition. In the absence of specific data, it is the practice in many risk assessments to simply use the observations from the Beyer et al. (1994) analysis and assume a soil ingestion rate for species located at the site of interest based on a closely related species that is described in Beyer et al. (1994). Commonly, species occurring at sites of interest are not closely related to those described by Beyer et al. (1994). As a result, if the soil ingestion pathway is a dominant contributor to total exposure, this may be a very significant source of uncertainty in risk assessments and subsequently in remedial goals that may be developed.

The analytical approach used by Beyer et al. (1994) for estimating soil ingestion is not especially difficult nor complex, yet yields data that are critical to accurately represent bird and mammal exposure. It is recommended that site-specific measurement of soil ingestion, following the method from Beyer et al. (1994) be included as a regular component of BERA data collection for the key bird and mammal receptors. These data will not only improve the resolution of the BERA conclusions but will also be available for development of Eco-SCVs in support of remediation.

Spatial exposure modeling

The approach for calculating the Eco-SSL uses an AUF of 1, which means that consumer organisms are assumed to spend 100% of their time foraging at the contaminated site. The REACH secondary poisoning assessment also simplifies the extent of foraging at a given site by assuming a 50%:50% split between the contaminated and uncontaminated areas. Both of these approaches can be refined with currently available information and techniques.

A Society of Environmental Toxicology and Chemistry (SETAC) Pellston Workshop on population risk assessment (Barnthouse et al. 2007) serves as the basis for discussion of how to define the foraging ranges of consumer organisms within the site of interest (the "assessment population") and describes approaches for extrapolating from individual-based exposure (as represented by the standard exposure model) to population-level effects. Whether or not metals in soils pose risks to wildlife species depends on the defined assessment population and several overlapping spatial scales relevant to metal exposures to this assessment population. Relevant spatial scales can include those related to the distribution of metals, the locations and arrangements of terrestrial habitats, the distribution and ecology of the assessment population, and imposed political or site boundaries that define an assessment area. For example, Carlsen et al. (2004) recommend integrating the spatial extent of contamination with receptor-specific home range or critical patch size. Similarly, Ryti et al. (2004) proposed the use of a population area use factor (PAUF), which is the proportion of the area required for the assessment population of the selected receptor (defined using statistical relationships between receptor-specific dispersal and home range size) that is represented by the contaminated site.

The goal of an Eco-SCV is to delineate a spatial area for which soil–metal concentrations exceed acceptable limits. To this end, the assessment populations are the wildlife species that have been identified as being relevant in the problem formulation step of the assessment. The relationship between the range of the species with the spatial area of the site that is being assessed will determine whether or not adjustment of the AUF is warranted, and to some degree, how important this adjustment will be. For example, it may not be relevant to modify the AUF for species with small foraging ranges that are being assessed for spatially large sites, as it may be reasonable to expect that the population spends the majority of its time foraging on the contaminated site. On the other hand, for species with large foraging ranges that are being assessed for small sites, it might be reasonable to expect that the population spends the

majority of its time foraging off of the contaminated site and assign a very small AUF. AUFs less than 1 should be applied at the screening stage when reasonable to do so. In some cases, this will yield a cost-effective "acceptable risk" conclusion without expending considerable resources trying to quantify other uncertainties that are considerably more difficult to assess.

Exposure experienced by individual animals and the population as a whole will depend on the integration of the distribution and ecology of individuals and the spatial distributions of metals contamination. Although there are many retrospective approaches available for investigating exposures, prospective approaches are of greatest interest here and typically involve some form of spatial modeling, for which several examples exist. Two such models are the SEEM (Wickwire et al. 2004) and Eco_SpaCE (Loos et al. 2010).

SEEM was developed for the US Army as a means for assessing the ecological risks of metals and other contaminants in soils to defined assessment populations. SEEM offers the risk assessor the opportunity to improve the ecological relevance of the risk assessment by considering spatial aspects of exposure through an evaluation of heterogeneous habitat use and chemical distribution, and a comparison of exposure with the potential for toxicological effects, resulting in a measure of risk to the population on site. SEEM predicts and compiles exposures for all individuals within a user-defined local population, rather than a single representative individual. In addition, SEEM increases the predictive capabilities of the exposure assessment by incorporating habitat preferences in the determination of daily exposure estimates. The model tracks an individual over an ecologically relevant period of time as it travels across a landscape. The individual moves according to a set of predetermined rules and exposure for a population of individuals can be tracked over time. The module was developed for inclusion within the Adaptive Risk Assessment Modeling System (ARAMS). The model follows a simple set of rules and can be used to estimate exposures. Work has also occurred to field-validate the model (Johnson et al. 2007). The model could be used in "what-if" contaminant scenarios to help establish exposure regimes—metals in soils—that would be protective of specific wildlife species. These "what if" scenarios could be run as iterations to develop tables that incorporate spatial features and dimensions. This may be a useful analysis to test the assumptions made under the REACH secondary poisoning assessment, which for local sites assumes that consumer organisms obtain 50% of their exposure from local sites and 50% from regional background.

Eco SpaCE (Loos et al. 2010) was developed to assess the combination of chemical, biological, and physical stressors on select wildlife species. Eco-SpaCE is a receptor-oriented cumulative exposure model for wildlife species that includes relevant ecological attributes such as spatial habitat variation, food web relations, predation, and life history characteristics. The model has been illustrated with a case study in which the predicted mortality due to Cd contamination is compared with the predicted mortality due to flooding, starvation, and predation for 3 small mammal species (wood mouse [Apodemus sylvaticus], common vole [Microtus arvalis], and European mole [Talpa europaea]) and a predator (little owl, Athene noctua) living in a lowland floodplain along the river Rhine in the Netherlands. It is likely that Eco SpaCE could also be run in an iterative fashion to explore the implications of different exposure regimes for metals.

Tissue-based exposure estimation

Evaluation of risks to wildlife has predominantly focused on estimation of dietary exposure that is then compared to literature-derived effect concentrations. This approach maximizes use of available data, but requires that the model address contaminant bioavailability, habitat use, and food use to accurately represent exposure. Because toxicity is a function of exposure at the organ-level, tissue-based assessments represent an alternative approach for exposure estimation that sidesteps dietary exposure estimation uncertainties and provides a direct exposure metric that can be more explicitly linked to toxic effects. Target-organ based exposure models may be either empirical (based on analyses of site-specific relationships between concentrations in soil and those in target organ tissues) or mechanistic (i.e., physiologically based toxicokinetic [PBTK] models).

Empirical target-organ-based exposure models have been developed and applied at multiple sites. One example is the CdA River Basin (USEPA 2001) for which target-organ-based exposure models were developed for Pb for waterfowl, an aquatic songbird, small mammals, and riparian songbirds. The CdA waterfowl model was developed using site-specific information and an adaptation of the exposure and/or effects model presented in Beyer et al. (2000). This model was used to estimate concentrations of Pb in blood and liver of tundra swans, Canada geese, mallards, and wood ducks resulting from incidental ingestion of sediment. The exposure model is of the form:

$$C_{Pb} = e^{[slope \times \ln[C_s \times (b - y + ay)/(ay - c + b)] + intercept]},$$

where C_{Pb} = estimated concentration of Pb in blood or liver (mg/kg wet weight) (separate estimates were generated for tundra swan, Canada goose, mallard, and wood duck); a = proportion digestibility of diet; b = proportion of acid-insoluble ash in diet; c = proportion of acid-insoluble ash in sediment; y = proportion of acid-insoluble ash in scat; C_s = concentration of Pb in sediment (mg/kg dry weight); slope = slope from species-specific diet-to-blood or diet-to-liver regression models; and intercept = intercept from species-specific diet-to-blood or diet-to-liver regression models.

Previous research from the CdA basin has indicated exposure of waterfowl to Pb through the food pathway is trivial compared to exposure from incidental sediment ingestion (Beyer et al. 2000). Therefore, oral or dietary exposure is equivalent to sediment exposure. Using the estimated concentrations of Pb in blood or liver, diet-to-blood and diet-to-liver bioaccumulation models were developed. Site-specific data from studies in which waterfowl were fed diets containing sediment from the CdA basin and bioaccessibility was measured (Heinz et al. 1999; Hoffman et al. 2000; Day et al. 2003) were also incorporated in the model. Tissue concentrations estimated using the site-specific models closely mirrored that measured in field collected birds (USEPA 2001). Additional models for an aquatic songbird and small mammals were developed and applied as part of the CdA ERA (USEPA 2001). Models for riparian songbirds in the CdA Basin were developed and presented in Sample et al. (2011).

For some metals such as Cd and Hg, certain organs are recognized as primary sites of accumulation. For example, the kidney is known to be the target organ for Cd toxicity. When kidney Cd concentrations in laboratory mammals exceeds

100 mg/kg, overt signs of tissue damage and resulting impairment of renal function may be expected (Nicholson et al. 1983; Friberg 1984). This knowledge supports the development of PBTK models to predict the uptake and distribution of metals accumulated to target organs and the resulting potential for tissue damage.

In PBTK modeling, the individual organism is modeled as a series of different compartments that represent tissues and/or organs that are connected by the circulation system. The chemical is assumed to enter through a range of routes including ingestion, dermal contact, and inhalation. Once absorbed, the chemical reaches the tissue compartments by blood circulation where it can accumulate as a result of its rate of entry through arterial blood and the rate of exit by venous blood. To enter the organ compartments, chemicals must cross biological membranes into the organ itself where it can accumulate. PBTK approaches have been developed for humans and are already extensively used in drug development and drug safety research (Rowland et al. 2011) and have also been adapted to metals (Ruiz et al. 2010). PBTK models developed for rodents for use in toxicological analysis are well established and have obvious potential for application to wildlife exposure (Timchalk et al. 2002). In ecotoxicology, there have been recent steps taken toward increasing the availability of models for wildlife species. Examples include the application of PBTK models to assess the role of organochlorines in reproductive toxicity for polar bears (Sonne et al. 2009), models and model design to simulate avian pesticide exposure in domestic fowl and gamebirds (Cortright et al. 2009), and promisingly in the context of the ecological risk assessment of metal for methylmercury in kestrels (Nichols et al. 2010).

A driver behind the growing interest in PBTK modeling is that the conceptual models are designed so that they represent the major processes involved in chemical adsorption, distribution, metabolism, and excretion. To optimize the model for concentration prediction in represented organs, it is important that relevant understanding and information on physiology, anatomy of the organism, and the physicochemical properties of the studied chemical are used. The physiological parameters that required are often well known for humans and model laboratory species, such as rats and mice. For wildlife values for parameterization may not be readily available, however, this issue can be addressed in part using allometric scaling techniques to derive parameter estimates (Young et al. 2001). Although allometric scaling may be a first step in applying physiological parameters to wildlife species, the resulting estimates will be uncertain.

Identification of vulnerable target organs and prediction of external concentrations can help focus biomonitoring studies on relevant endpoints. With PBTK models increasingly available there is the potential to combine model prediction to monitoring studies for environmentally relevant small mammal species using biomarker approaches. For example Griffin et al. (2000) have shown the effects of As at environmentally relevant exposure concentrations on voles identifying a number of metabolic changes that can provide tools for biomonitoring and assessment of organ toxicity in the field.

Overall, if tissue-based effect thresholds are available for the receptors and contaminants for which risks are identified, empirical or mechanistic tissue-based exposure models should be considered as an additional exposure metric for derivation of EcoSCVs. If suitable mechanistic models are lacking, site-specific field data may be collected such that empirical, site-specific models could be developed.

PROPOSED APPROACH FOR INTEGRATING IMPROVED EXPOSURE ESTIMATION FOR ECO-SCV DERIVATION

As the previous sections indicate, the incorporation of realism serves to reduce the uncertainty in site-specific ecological risk assessment for metals in soils and also for the derivation of Eco-SCVs. These reductions in uncertainty typically occur as part of a phased approach that begins with screening values such as Eco-SSLs and proceed to include more site-specific and receptor-specific information on exposure and effects. Phased approaches are commonly used in risk assessments as a means to balance resources against the desire to reduce uncertainty in the assessments. This process is reflected in the recommendations made in this article. Examples of phased risk-based approaches include the USEPA Superfund Program (USEPA 1997), the ASTM Risk-Based Corrective Action (RBCA) standard for ecological assessments and site closure (ASTM 2002), and approaches taken by many states such as California, (DTSC 1996), Oregon (ODEQ 1998), Washington (WADOE 2001), and Texas (TCEQ 2006). With the prevalence of phased approaches to risk assessment, it makes sense to use an analogous approach for incorporating realism into the derivation of Eco-SCVs. The RBCA approach can serve as a template for the incorporation of site-specific information to reduce uncertainty and to derive Eco-SCVs. These refinements to exposure estimates for wildlife should be carried out as needed during the remedial investigation phase and those refinements should then be incorporated into the development of site-specific Eco-SCVs. As with recent USEPA guidance on considering bioavailability of soil contaminants with respect to human exposures (USEPA 2007c), the process of incorporating such information into the derivation of Eco-SCVs should take into account the value of such additional information for decision making.

The mechanics of deriving an Eco-SCV flow from the same equations used to estimate exposure and risk to wildlife but with a slight rearrangement. Realism in site-specific wildlife Eco-SCVs can be achieved by obtaining more realistic estimates for diet composition, bioaccumulation, bioavailability and/or bioaccessibility, soil ingestion, spatial aspects of exposure, and target organ exposure. These components of wildlife exposure estimation should be developed on a site-, species-, and analyte-specific basis to the extent that the expense for their derivation is justified by the value they add to Eco-SCV development.

As suggested in previous sections, there are a number of tools and approaches available that can improve realism and reduce uncertainty and associated conservatism to arrive at appropriate cleanup goals. However, deriving Eco-SCVs related to wildlife should follow a set of risk-management principles outlined below:

- As stated in USEPA's Framework for Metals Risk Assessment, metals occur naturally in the environment and the ambient levels of metals vary with soil type and with geography. Eco-SCVs should not be less than and can be greater than ambient conditions for the area of interest. If modeled approaches are yielding Eco-SCVs that are at or less than ambient, then it should be presumed that the analysis lacks sufficient realism for the area and receptors of

interest. At that point, either the risk assessment should be taken to the next phase, or if that is cost prohibitive, then adaptive management should be used to address the unresolved uncertainty about risk.

- Metal exposures to wildlife involve estimating exposures via: 1) incidental soil ingestion, and 2) food items that have bioaccumulated metals from soils. The use of Eco-SCVs can be improved by incorporating site-specific realism into one or both of these 2 exposure pathways.
- Sensitivity analyses can be used to evaluate the value of collecting additional information to reduce the uncertainties in metals exposure estimates for wildlife. These can provide insight into whether exposure is related to soil ingestion, bioaccumulation, or a combination of pathways. The value of collecting additional information is related to what factors underpin site management decisions. For example, management of risks could be influenced by other chemicals or other receptors such as humans. In such cases, there is lower value associated with improving on wildlife exposure estimates. However, if management decisions rest on wildlife exposures, there is high value in improving on exposure estimates by incorporating site- or region-specific information.
- Toxicity reference values and exposure estimates must be properly aligned to avoid errors in estimating risks and deriving soil target levels. For example, if effects are based on critical body residues, then exposure estimates must provide such estimates; if they are based on oral doses or bioaccessible fractions, then exposure estimates must deliver that information. Mayfield et al. (2013) provide recommendations for improved derivation of wildlife toxicity data to supported Eco-SCV development.
- The degree of bioaccumulation of metals by soil invertebrates and plants depends on the form of the metal as well as the characteristics of the soils. Models and tools are available for making estimates that consider these factors and these site- and soil-specific models are preferable to default approaches used to derive screening levels (e.g., use of generic BAFs). Site-specific relationships can also be developed from measurements of soils and biota and these relationships are generally preferable to modeled tissue levels. The selection of a modeling or measurement approach will depend on available information, methods, and the relative costs associated with obtaining the information relative to the value of that information for decision making.
- Whereas the soil metal concentrations at sites under evaluation are undoubtedly elevated and represent an important source of exposure, the bioaccessibility and bioavailability of metals in soils may be less than in water and food. Therefore, presuming that bioaccessibility and/or bioavailability is equivalent among these sources may result in an overestimate of metals exposure to wildlife species that incidentally ingest soils. Models and tools are available for developing more accurate and more certain estimates of metals exposures for wildlife that incidentally ingest soils.
- The derivation and application of Eco-SCVs should be at appropriate spatial scales. This can be accomplished by considering the spatial scale of the wildlife receptors in relation to the spatial scale of metal contamination. AUFs refer to areas over which the assessment population forages and include areas for individuals as well as the overall assessment population. These areas can overlap to varying degrees with the areas containing elevated levels of metals associated with anthropogenic sources. Exposures can be evaluated and Eco-SCVs derived that are judged to be protective for the assessment population (e.g., set as a percentage of the population). Eco-SCV values may be separately derived that are protective for acute exposures as well as chronic exposures.
- Regardless of the methods used for exposure estimation, implementation of remedial programs based on derived Eco-SCVs should consider all of the factors used to judge the efficacy as well as the short- and long-term consequences of alternative remedial actions. From an ecological standpoint, remedial alternatives should strive to achieve optimum ecological benefits associated with risk reduction goals. In some cases, this may involve innovative solutions.

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SUPPLEMENTAL DATA

Ni secondary poisoning sensitivity analysis

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